

An analytical solution to contaminant advection and dispersion through a GCL/AL liner system

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An one-dimensional model for contaminant advection and dispersion through a GCL/AL liner system was developed to analyze the equivalence between a GCL (geosynthetic clay liner) and a CCL (compacted clay liner). The continuity of contaminant concentration and flux at the interface between the GCL and the underlying attenuation liner (AL) are obeyed in the model, and background concentrations in the soil liner are also considered. Based on the assumption that contaminant transport through a GCL was a steady state process, an analytical solution was obtained. Increasing the leachate head from 0.3 m to 10 m results in a reduction of the breakthrough time of benzene by a factor of 2.7. The breakthrough time of benzene increases by a factor of 7.0 when the hydraulic conductivity of GCL decreases by one order of magnitude. The breakthrough curves are more sensitive to the hydraulic conductivities of the GCL and AL (attenuation layer) than to the thickness of the AL. The standard 75 cm CCL can be replaced by a combination of a GCL and a 1.0–4.0 m thickness of AL. The proposed method can be used for preliminary design of GCL composite liners, assessing the equivalence between GCL and CCL, preliminary design of a remediation method for contaminated soils, and evaluating experimental results.

GCL, compacted clay liner, attenuation layer, leachate, advection-dispersion, equivalence analysis

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Globally, 1.2 billion people lack access to safe drinking water and about 2.6 billion have little or no sanitation services provided [1,2]. Groundwater is one of the important sources of drinking water. In the past 30 years, groundwater supply has satisfied 20% of the total water requirements in China and 52% in the northern water deficit area [3]. In the U.S., approximately 50% of the drinking water supply originates from groundwater [4]. Municipal solid waste landfills have been a great threat to groundwater contamination [5,6]. Landfilling is by now the major treatment method for municipal solid wastes in China [7,8]. Therefore, the potential for groundwater contamination caused by landfilling requires assessment by mathematical models.

To protect underlying groundwater resources from landfills, waste disposal sites are commonly lined with multi-layered liner systems [9, 10]. Geosynthetic clay liners (GCL) have been widely used in landfill liner systems to replace the traditional compacted clay liners (CCL) because of their low price, easily controlled construction quality and low permeability [11–13]. The Chinese standard (CJJ 113-2007) includes a provision that permits the use of GCL composite liners, provided that an attenuation layer (AL) of a certain thickness is added beneath the GCL [14]. A GCL is usually combined with an AL in many landfills in developed countries [15–17]. The permeability of the AL is required to be $< 1 \times 10^{-7}$ cm/s in the Chinese standard, but the thickness of the AL is not specified [14]. In France, the thickness of the AL beneath the GCL should be > 0.5 m [17].

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A mathematical model for contaminant advection-dispersion-adsorption through the GCL/AL system is needed to assess and predict the performance of the GCL/AL. Mathematical models for contaminant advection-dispersion through layered media have usually been solved numerically because of their complicated nature. Leo and Booker developed a contaminant transport model for non-homogeneous media using a boundary element method [18]. Guyonnet et al. [17] and Zhan et al. [19] studied the hydro-dispersion equivalence between multi-layered mineral layers. Rowe et al. [15] studied the problem of contaminant advection-dispersion in layered soils by the finite-layer method. Zhang et al. [20] developed a model for contaminant transport through a three-layered landfill bottom media by the finite difference method. Cooke and Rowe [21] used the finite element method to study two-dimensional contaminant transport through a landfill bottom barrier system.

Analytical solutions have irreplaceable advantages over numerical methods. Analytical solutions can be used to verify the accuracy, convergence and stability of various computational methods and programs, and can also be used as a standard solution to stimulate the development of various numerical methods [22]. The analytical solutions are more suitable for preliminary design of landfill liners and general decision-making than the numerical models [23, 24].

Foose et al. [25] has given the analytical solution for one-dimensional diffusion through a composite liner with a semi-infinite bottom boundary. Chen et al. [26] and Xie et al. [27] have provided the analytical solutions for contaminant diffusion through multi-layered media for different boundary conditions. Huysmans and Dassargues [28] developed the equivalent diffusion coefficient model for contaminant diffusion through multilayered media. However, the advection and dispersion effects on contaminant transport through layered media caused by the hydraulic gradient were not considered in these studies. In many developing countries, the leachate head generated in landfills tends to be very high because of their low construction quality (e.g., the leachate head exceeds 10 m in the Qizishan landfill in China) [5, 6, 29, 30]. Under this circumstance, the effects of advection and dispersion can be of great importance and cannot be ignored. It is very difficult to obtain the analytical solutions for considering the effect of advection and dispersion on contaminant transport through layered media [23] and analytical solutions for contaminant transport through GCL/AL are not available.

In this paper, steady state contaminant transport through a GCL was assumed and concentration continuity and flux continuity between the GCL and AL was introduced to develop a one-dimensional model for contaminant advection-dispersion through a GCL/AL. The Laplace transformation was used to obtain dimensionless analytical solutions in terms of concentration and flux. Detailed analysis of the proposed analytical solutions was carried out. The equivalence between GCL and CCL was also analyzed. The

proposed analytical solution can be used for the prediction of contaminant transport through a GCL/AL system, preliminary design of landfill GCL/AL liner systems, verification of more sophisticated numerical models and evaluation of experimental results.

1 Analytical solution

The top layer of the landfill liner system is assumed to be a GCL. Because GCLs are very thin, contaminant transport through this layer can be assumed to be a steady state process. The governing equation for contaminant transport through GCL is

$$D_r \frac{d^2 C_r}{dz^2} - v_r \frac{dC_r}{dz} = 0, \quad (1)$$

where C_r is the pore water concentration of the contaminant in the GCL; v_r is the seepage velocity of the GCL; and D_r is the hydrodynamic coefficient of the GCL. D_r can be determined from

$$D_r = D_r^* + D_{m,r}, \quad (2)$$

where D_r^* is the effective diffusion coefficient of the GCL and $D_{m,r}$ is the mechanical dispersion coefficient of the GCL. Conservatively, $D_{m,r}$ can be determined by [31]

$$D_{m,r} = \frac{L_r}{10} v_r, \quad (3)$$

where L_r is the thickness of the GCL.

The transport of contaminants through the soil liner beneath the GCL can be given by the transient advection-dispersion equation:

$$\frac{\partial C_A(z,t)}{\partial t} = \frac{D_A}{R_d} \frac{\partial^2 C_A(z,t)}{\partial z^2} - \frac{v_A}{R_d} \frac{\partial C_A(z,t)}{\partial z}, \quad (4)$$

where $C_A(z,t)$ is the porewater concentration of the contaminant in the soil liner at any time t and any position z ; D_A is the hydrodynamic coefficient of the soil liner; R_d is the retardation factor of the soil liner; and v_A is the seepage velocity of the soil liner.

$D_{m,A}$ can be determined by

$$D_A = D_A^* + D_{m,A}, \quad (5)$$

where D_A^* is the effective diffusion coefficient of the AL and $D_{m,A}$ is the mechanical dispersion coefficient of the AL. As for the GCL, $D_{m,A}$ can be determined by

$$D_{m,A} = \frac{L_A}{10} v_A. \quad (6)$$

The retardation factor R_d can be determined by

$$R_d = 1 + \frac{\rho_b K_d}{n_A}, \quad (7)$$

where ρ_b is the dry density of the soil; K_d is the distribution coefficient of AL; and n_A is the porosity of the AL.

A schematic diagram of contaminant advection-dispersion through the liner system of a GCL/AL is shown in Figure 1. The contaminant concentration in the leachate is assumed to be a constant C_0 .

The top boundary condition of this double liner system can then be expressed as follows:

$$C_r(0) = C_0. \quad (8)$$

The background concentration in the AL is assumed to be C_i . The initial condition for governing eq. (4) can then be written as

$$C_A(z, 0) = C_i. \quad (9)$$

The continuous conditions between GCL and AL can be expressed as [15]

$$C_r(L_r) = C_A(L_r, t), \quad (10)$$

$$n_r D_r \frac{\partial C_r(L_r, t)}{\partial z} = n_A D_A \frac{\partial C_A(L_r, t)}{\partial z}, \quad (11)$$

$$n_r v_r = n_A v_A = v_a, \quad (12)$$

where n_r is the porosity of the GCL and v_a is the Darcy velocity of the double liner system which can be determined by

$$v_a = \frac{h_w + L_{\text{sys}}}{L_{\text{sys}}} \times K_{\text{sys}}, \quad (13)$$

where L_{sys} is the thickness of the GCL/AL; K_{sys} is the hydraulic conductivity of the system; and K_{sys} is the harmonic mean of the GCL and AL:

$$K_{\text{sys}} = \frac{L_r + L_A}{\frac{L_r}{K_r} + \frac{L_A}{K_A}}, \quad (14)$$

where K_r is the hydraulic conductivity of the GCL and K_A is the hydraulic conductivity of the AL.

The bottom boundary of the AL is assumed to be semi-infinite, and then the bottom boundary condition for the problem is

$$\frac{\partial C_A(\infty, t)}{\partial z} = 0. \quad (15)$$

The analytical solution for governing eq. (4) satisfying all solving conditions is (see Appendix for the derivation of the solution)

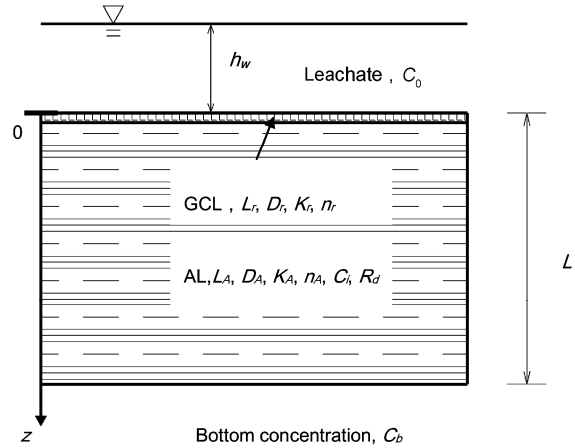


Figure 1 Schematic diagram of contaminant transport through the GCL/AL.

$$\frac{C_A(P_{LA}, T) - C_i}{C_0 - C_i} = \frac{1}{2} \left\{ \operatorname{erfc} \left(\frac{1-T}{2\sqrt{T/P_{LA}}} \right) + e^{P_{Lr} + P_{LA}} \operatorname{erfc} \left(\frac{1+T}{2\sqrt{T/P_{LA}}} \right) - e^{\frac{P_{LA}}{4}(-1+e^{P_{Lr}}+T)\operatorname{csch}^2\left(\frac{P_{Lr}}{2}\right)} (1+e^{P_{Lr}}) \operatorname{erfc} \left[\frac{1+T \coth\left(\frac{P_{Lr}}{2}\right)}{2\sqrt{T/P_{LA}}} \right] \right\}, \quad (16)$$

where

$$P_{Lr} = \frac{v_r L_r}{D_r}, \quad (17)$$

$$P_{LA} = \frac{v_A z}{D_A} \quad (L_r \leq z \leq L), \quad (18)$$

$$T = \frac{v_A t}{R_d z} \quad (L_r \leq z \leq L). \quad (19)$$

The flux at the bottom of the liner system of the GCL/AL, J_A , can be obtained by

$$J_A = n_A v_A C_A(L, t) - n_A D_A \frac{\partial C_A(z, t)}{\partial z} \Big|_{z=L}. \quad (20)$$

Substituting eq. (16) into the above equation results in

$$\frac{J_A - n_A v_A C_i}{n_A v_A (C_0 - C_i)} = \frac{1}{2} \left\{ \operatorname{erfc} \left(\frac{1-T}{2\sqrt{T/P_{LA}}} \right) - e^{\frac{P_{LA}}{4}(-1+e^{P_{Lr}}+T)\operatorname{csch}^2\left(\frac{P_{Lr}}{2}\right)} \coth\left(\frac{P_{Lr}}{2}\right) \operatorname{erfc} \left[\frac{1+T \coth\left(\frac{P_{Lr}}{2}\right)}{2\sqrt{T/P_{LA}}} \right] \right\}. \quad (21)$$

When the thickness of the GCL (L_r) in eqs. (16) and (17) approaches zero, the proposed analytical solution [eq. (16)] can be reduced to the analytical solution for contaminant transport in homogeneous soil:

$$\frac{C_A(P_{LA}, T) - C_i}{C_0 - C_i} = \frac{1}{2} \left\{ \operatorname{erfc} \left(\frac{1-T}{2\sqrt{T/P_{LA}}} \right) + e^{P_{LA}} \operatorname{erfc} \left(\frac{1+T}{2\sqrt{T/P_{LA}}} \right) \right\}. \quad (22)$$

The above equation is Ogata's classical analytical solution [32, 33]. The result indicates that the proposed solution is reasonable.

2 Values for transport parameters

Cd^{2+} and benzene were chosen to represent inorganic and organic constituents in the leachate, respectively. The transport parameters of the GCL, CCL, and AL for the two contaminants are shown in Table 1. The parameters were obtained from Rowe et al. [15].

The concentration of Cd^{2+} was assumed to be 1 mg/L, which is a typical value in Chinese landfills [34]. The benzene concentration in the leachate was assumed to be 1.63 mg/L [35]. Both of the allowable concentrations of Cd^{2+} and benzene in groundwater are 0.005 mg L⁻¹, which is based on the US EPA standard for drinking water [36].

3 Parametric studies

For the GCL/AL, parametric studies were carried out to investigate the effect of leachate head, hydraulic conductivity and thickness of the AL on the breakthrough curves of the liner system. The liner system is assumed to be a combined GCL and 5m thick AL. The leachate head is assumed to be 0.3 m. The other parameters are the same as those shown in Table 1.

3.1 Effect of leachate head

The effect of leachate head on benzene breakthrough curves

is shown in Figure 2. The reference case was assumed to have $h_w = 0.3$. This leachate head is also the limit value specified by the US standard [10]. The leachate head greatly influenced the breakthrough time and bottom flux of benzene. When the leachate head increases from 0.3 m to 10 m, the breakthrough time of benzene decreases by a factor of 2.7 (Figure 2(a)) and the 100-year flux at the bottom of the liner system increases by a factor of 2.8 (Figure 2(b)). The breakthrough time and 100-year bottom flux for $h_w = 0.3$ m are very similar to that for $h_w = 1$ m with the differences for breakthrough time and 100-year bottom being 10%, and 12%, respectively. This indicates that the landfill liner system would provide an effective barrier when the leachate

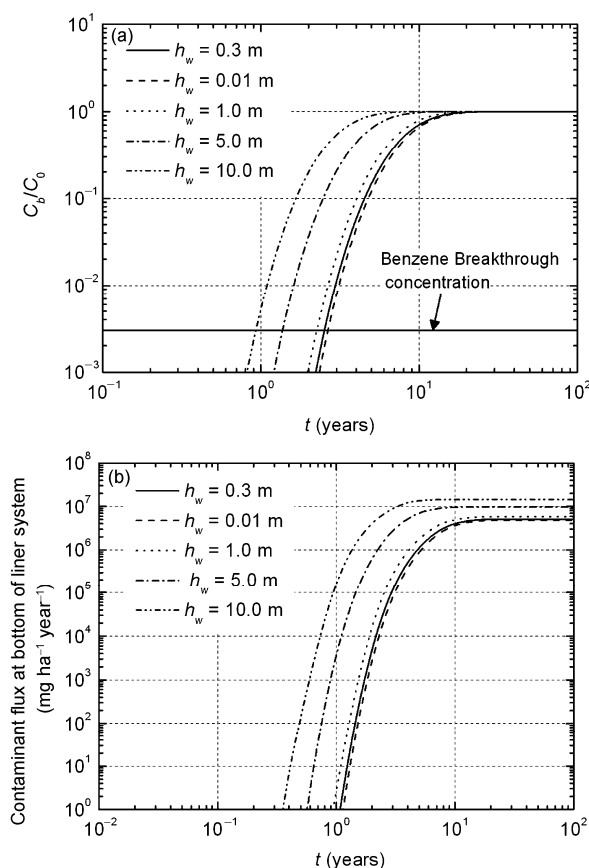


Figure 2 Effect of leachate head on the breakthrough curves of GCL/AL. (a) Concentration; (b) flux.

Table 1 Transport parameters for Cd^{2+} and benzene in liners

Parameters		CCL	GCL	AL
Thickness (m)		0.75	0.0138	1~10
Porosity		0.54	0.86	0.40
Hydraulic conductivity (m s ⁻¹)		1.0×10^{-9}	5.0×10^{-11}	1.0×10^{-7}
Dry density (g cm ⁻³)		1.79	0.79	1.62
Effective coefficient (m ² s ⁻¹)	Cd^{2+}	1.76×10^{-10}	3.6×10^{-10}	8.9×10^{-10}
	benzene	5.0×10^{-10}	3.3×10^{-10}	8.9×10^{-10}
	Cd^{2+}	0.36	—	0.36
K_d (mL g ⁻¹)	Cd^{2+}	0.28	—	0.28
	benzene	0.28	—	0.28

head is controlled to be less than 1 m, but when the leachate head reaches 10 m, the performance of the liner system decreases greatly. Increasing the leachate head from 0.3 m to 10 m reduces the breakthrough time of benzene by a factor of 2.8 and increases the 100-year bottom flux by a factor of about 3. Therefore, it is of great importance to restrict the leachate head to a relatively low value (e.g. <1 m).

3.2 Effect of the hydraulic conductivity of GCL

The effect of the hydraulic conductivity of GCL on benzene breakthrough curves is shown in Figure 3. According to reference [11], the hydraulic conductivity of GCL range from 10^{-12} to 10^{-10} m s⁻¹. A hydraulic conductivity of 5×10^{-11} m s⁻¹ was chosen as the reference case. This reference value is the maximum hydraulic conductivity of GCL specified by the Chinese standard [14]. The results indicate that the hydraulic conductivity of GCL has a significant influence on the benzene breakthrough time and bottom flux. Increasing the hydraulic conductivity of a GCL by a factor of 5 results in a reduction of the breakthrough time by a factor of 1.7 and an increase in the 100-year contaminant flux at the bottom of the liner system by a factor of 1.7. Decreasing the hydraulic conductivity of the GCL by a factor of 10 increases the breakthrough time by a factor of 7 and decreases the 100-year contaminant flux at the bottom of the liner system by up to an order of magnitude. Decreasing the hydraulic conductivity of the GCL to 1×10^{-12} m s⁻¹, increases the breakthrough time by a factor of 20.7 and the 100 year bottom flux decreases by up to 5.6 orders of magnitude. The hydraulic conductivity of the GCL therefore plays an important role in the performance of the GCL/AL liner system. High quality construction of the GCL should be ensured to restrict the hydraulic conductivity of the GCL to a relatively low level (e.g. $<1 \times 10^{-12}$ m s⁻¹).

3.3 Effect of the hydraulic conductivity of the AL

The hydraulic conductivity of the AL in the field tends to change greatly because control of the construction quality of clay soils is very difficult. The hydraulic conductivity of the AL should be less than 1×10^{-7} m s⁻¹ and the hydraulic conductivity of compacted clay liner should be less than 1×10^{-9} m s⁻¹ [14]. The effect of the hydraulic conductivity of the AL on the performance of GCL/AL is shown in Figure 4. Similar to variations in the hydraulic conductivity of GCL, variation in the hydraulic conductivity of the AL greatly influences the performance of the GCL/AL. Assuming $K_A = 1 \times 10^{-7}$ m s⁻¹ to be the reference case, the breakthrough time increases by a factor of 2.3 and the 100-year bottom flux decreases by a factor of 2.4 when the hydraulic conductivity of the AL is decreased by an order of magnitude. The breakthrough time increases by a factor of 11.5 and the 100-year bottom flux decreases by a factor of about 30 when the hydraulic conductivity of the AL is decreased by 2

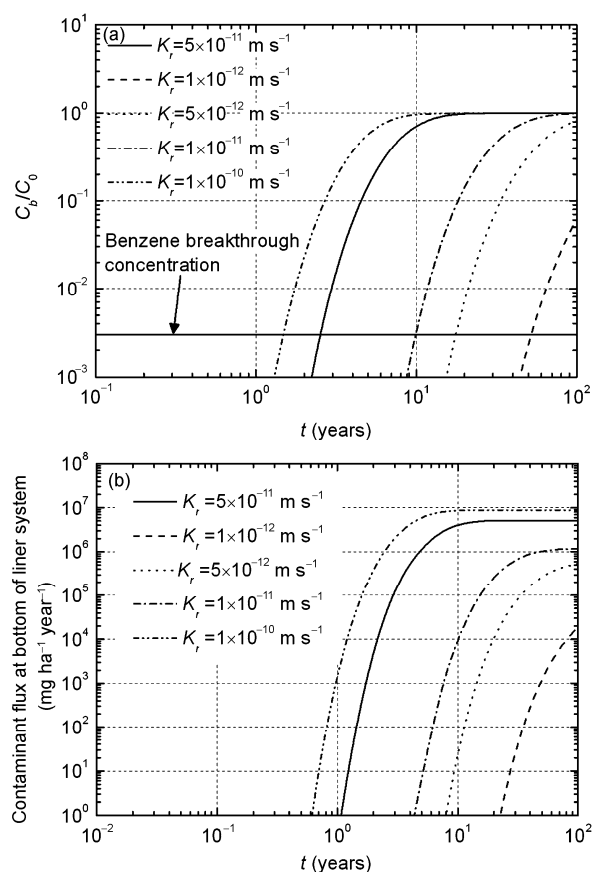


Figure 3 Effect of hydraulic conductivity of GCL on the breakthrough curves of GCL/AL. (a) Concentration; (b) flux.

orders of magnitude. In addition, the performance of the GCL/AL is not greatly affected when the hydraulic conductivity of the AL is increased by an order of magnitude with the breakthrough time only decreasing by 13% and the 100-year bottom flux only increasing by 16%. Comparisons with the results presented in Section 3.2 indicate that the breakthrough curves of GCL/AL are more sensitive to variation in the hydraulic conductivity of the GCL than to variation in the hydraulic conductivity of the AL. However, the performance of the liner system can be greatly improved by reducing the hydraulic conductivity of the AL to 1×10^{-9} m s⁻¹.

3.4 Effect of the AL thickness

Figure 5 shows the effect of AL thickness on the performance of the GCL/AL. The effect of the GCL layer on the performance of the liner system is also shown. Comparison of the breakthrough curves of a single 2 m AL and GCL+2 m of AL shows that the performance of the liner system is significantly affected by the GCL layer. The benzene breakthrough time of the GCL+2 m AL was 13.6 times greater than that of the 2 m AL alone and the 100-year bottom flux of the GCL+2 m AL was 14.7 times lower. For

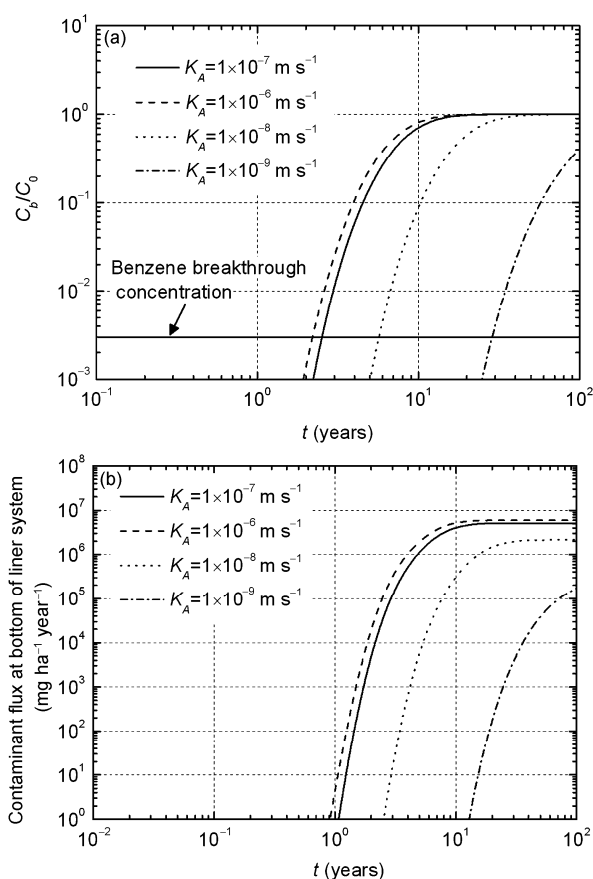


Figure 4 Effect of the hydraulic conductivity of the AL on the breakthrough curves of GCL/AL. (a) Concentration; (b) flux.

GCL/AL, the barrier performance was more sensitive to the decreased hydraulic conductivity of the GCL than to the increased thickness of the AL. The breakthrough time of the GCL+AL increases by a factor of 2.4 when the AL thickness is increased from 1 m to 10 m. Decreasing the hydraulic conductivity of the GCL by an order of magnitude increases the breakthrough time by up to a factor of 7. For breakthrough times, the protection level achieved by increasing the thickness of the AL by an order of magnitude is nearly the same as that achieved by decreasing the hydraulic conductivity of AL by up to an order of magnitude. However, the 100-year bottom flux tends to increase with increased AL thickness (Figure 5(b)). For example, increasing the AL thickness from 1 m to 10 m increases the 100-year bottom flux by a factor of about 6. This result is similar to those found by Shackelford [33], Foose et al. [25] and Rowe et al. [15], who indicated that the bottom flux increased over small and moderate time intervals and then decreased over longer periods of time.

4 Equivalent analysis between GCL and CCL

With the wide use of GCL in modern landfill sites, more

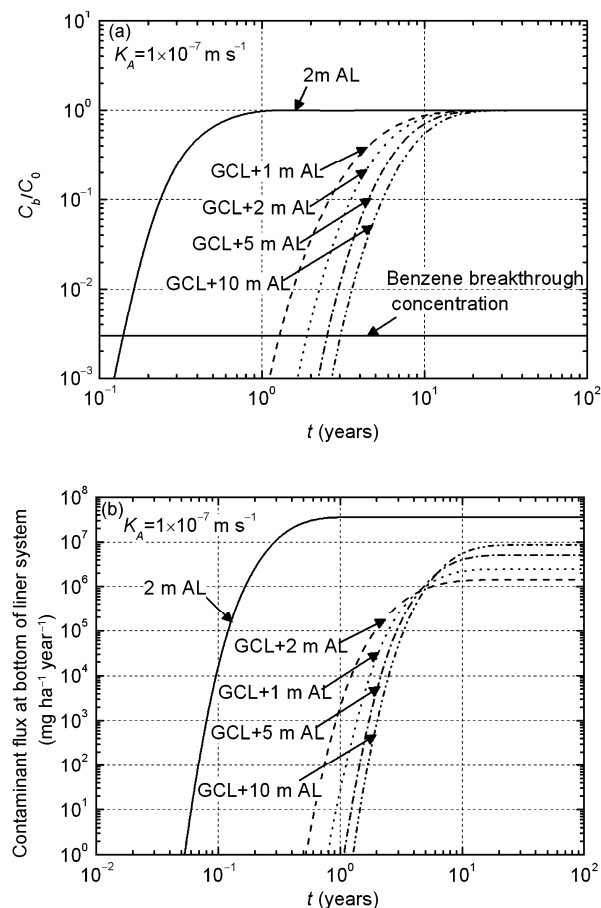


Figure 5 Effect of AL thickness on the breakthrough curves of GCL/AL. (a) Concentration; (b) flux.

and more importance has been attached to investigations of equivalence between GCL and CCL. In this study, the equivalence between GCL and CCL was investigated for Cd^{2+} and benzene. The required thickness of the AL beneath the GCL in order to achieve the same protection level as that given by the standard 75 cm CCL was analyzed. The leachate head was assumed to be 2 m. The different liner systems were considered to be equivalent when the breakthrough times were equal.

4.1 Equivalent thickness based on heavy metal ion breakthrough time

Three cases with different AL hydraulic conductivities of 1×10^{-6} , 1×10^{-7} and $1 \times 10^{-8} \text{ m s}^{-1}$ were considered. Variations in Cd^{2+} breakthrough time of the GCL/AL with the varying AL hydraulic conductivities are shown in Figure 6. The Cd^{2+} breakthrough time for a 75 cm CCL is also shown. To achieve the same breakthrough time as that for 75 cm of CCL, 1.3 m, 1.9 m and 2.1 m of AL in conjunction with the GCL is needed for $K_A = 1 \times 10^{-8} \text{ m s}^{-1}$, $K_A = 1 \times 10^{-7} \text{ m s}^{-1}$, and $K_A = 1 \times 10^{-6} \text{ m s}^{-1}$, respectively. There were only small differences between the breakthrough times for $K_A = 1 \times 10^{-7} \text{ m s}^{-1}$

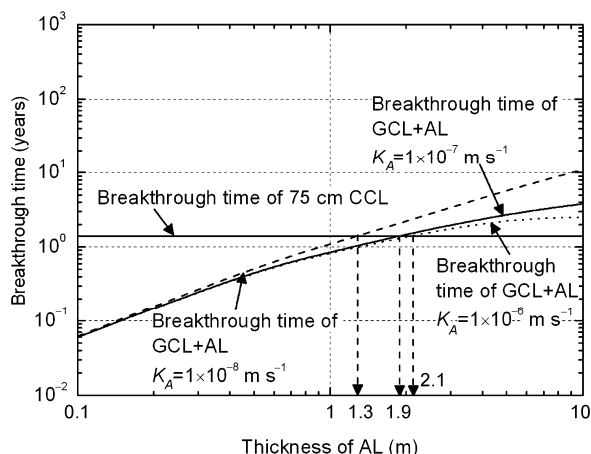


Figure 6 Equivalent thickness between GCL/AL and CCL for a metal ion.

and $K_A = 1 \times 10^{-6} \text{ m s}^{-1}$. However, the required thickness of the AL decreases by a factor of 1.5 with a decrease in the hydraulic conductivity of the AL from $1 \times 10^{-7} \text{ m s}^{-1}$ to $1 \times 10^{-8} \text{ m s}^{-1}$.

4.2 Equivalent thickness based on organic contaminant breakthrough time

As for the Cd^{2+} case discussed in the previous section, benzene breakthrough times for three different AL hydraulic conductivities of 1×10^{-6} , 1×10^{-7} and $1 \times 10^{-8} \text{ m s}^{-1}$ were considered. Variations in the benzene breakthrough times of the GCL/AL with these varying AL hydraulic conductivities together with the benzene breakthrough time for a 75 cm CCL are shown in Figure 7. To achieve the same breakthrough time as that for the 75 cm CCL, 1.5 m, 2.6 m and 3.2 m of AL in conjunction with the GCL is needed for $K_A = 1 \times 10^{-8} \text{ m s}^{-1}$, $K_A = 1 \times 10^{-7} \text{ m s}^{-1}$, and $K_A = 1 \times 10^{-6} \text{ m s}^{-1}$, respectively. As for Cd^{2+} , there were relatively small differences between the benzene breakthrough times for K_A

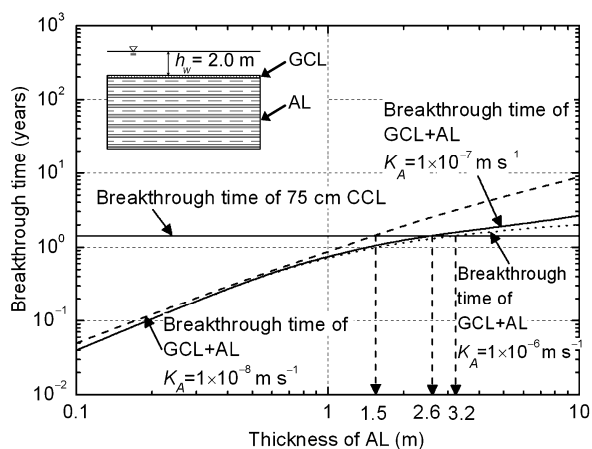


Figure 7 Equivalent thickness between GCL/AL and CCL for an organic contaminant.

$= 1 \times 10^{-7} \text{ m s}^{-1}$ and $K_A = 1 \times 10^{-6} \text{ m s}^{-1}$ (Figure 7) but the required thickness of AL decreases by a factor of 1.7 with a decrease in the hydraulic conductivity of the AL from 1×10^{-7} to $1 \times 10^{-8} \text{ m s}^{-1}$.

5 Conclusions

GCL/AL liners have been widely used in landfill systems. A one-dimensional advection-dispersion model for contaminant transport through GCL/AL was developed. Dimensionless concentration and dimensionless flux analytical solutions were obtained using a Laplace transformation. The performance of the GCL/AL system was evaluated on the basis of the analytical solution. The equivalence between GCL and CCL liner systems was also investigated. The main conclusions are as follows:

- (1) The proposed analytical solution can be reduced to the analytical solution for homogeneous soil. The proposed analytical solution can be used for verification of complicated numerical models, preliminary design and evaluation of GCL/AL liner systems, and for fitting experimental data.
- (2) The leachate head has a great influence on the performance of GCL/AL liner systems. Increasing the leachate head from 0.3 m to 10 m decreased the breakthrough time of benzene by a factor of 2.7 and the 100-year flux at the bottom of the liner system increased by a factor of about 3. It is of great importance that the leachate head acting on the GCL/AL system should be controlled to be less than 1 m.
- (3) The breakthrough curve of GCL/AL was more sensitive to variation in the hydraulic conductivity of the GCL than to variation in the hydraulic conductivity of the AL. The breakthrough time increased by a factor of 7 when the hydraulic conductivity of the GCL was decreased by 1 order of magnitude, while the breakthrough time increased by a factor of 2.3 when the hydraulic conductivity of the AL was decreased by the same factor. There were only small differences in breakthrough times between the case of $K_A = 1 \times 10^{-6} \text{ m s}^{-1}$ and $K_A = 1 \times 10^{-7} \text{ m s}^{-1}$.
- (4) A 75 cm CCL could be replaced by a combination of GCL and 1.0–4.0 m of AL for sites where a CCL is not available or where construction of a CCL is very difficult.

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Supporting Information

Table S1 PDB IDs for antigen-antibody complexes included in the testing dataset

The supporting information is available online at csb.scichina.com and www.springerlink.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.